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Occurrence of Fluoride in Drinking Water, Food, and Air (Draft)

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Prepared for

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OCCURRENCE OF FLUORIDE

IN

DRINKING WATER, FOOD, AND AIR

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16. ABSTRACT

The Office of Drinking Water (ODW), U. S. Environmental Protection Agency (USEPA) has prepared a draft document entitled Fluoride Occurrence in Drinking Water, Food, and Air. This Document is a preliminary draft which has not, as yet, been formally released by ODW, USEPA and should not be construed at this stage to represent Agency policy. This Document is a review of the available information on distribution of the fluoride occurrence levels in drinking water. The occurrence of fluoride in food and air are also reviewed. The Document provides estimates of the relative exposures from these sources.

17. KEY WORDS AND DOCUMENT ANALYSIS

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NOTICE

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SUMMARY

Fluoride is the generic term that is used in referring to compounds containing the element fluorine. Fluorides are ubiquitous in the human environment and their occurrence in drinking water, food, and air is influenced both by natural processes and human activities. However, except for the intentional addition of fluoride to drinking water for dental caries protection, its occurrence in drinking water, food, and air appears to be predominately the result of natural processes.

Fluorides are natural constituents of certain rock and mineral formations; the largest reserves of fluoride are found as the mineral fluorapetite in phosphate rock deposits. However, fluorspar, which contains a higher level of fluoride, is the primary commercial source of this element. Rocks and minerals appear to be the primary source of fluoride found in soils. Fluorides enter groundwater primarily by leaching from rocks and minerals. Fluoride enters surface water from soil runoff, industrial effluents, and deposition/precipitation of airborne particulates. Fluoride in lakes and rivers is deposited in sediments or is carried to the oceans where it is deposited in marine sediments.

The average level of fluoride in groundwater ranges from about 0.1 mg/l to more than 8 mg/l, depending on the type of rock with which it is associated. Surface water generally has fluoride levels averaging less than 0.3 mg/l.

Virtually all foods contain trace amounts of fluoride. However, few foods contain more than 1-2 ppm and most contain less than 0.5 ppm (dry weight).

Available data show that air levels of fluoride in suspended particulates generally do no exceed 0.05 ug/m^3, although higher levels in the vicinity of certain fluoride sources (e.g., near phosphate mining) have been observed.

Information on the occurrence of fluoride in public drinking water supplies is provided by three Federal surveys (1969 Community Water Supply Study, 1978 Community Water Supply Survey, and the Rural Water Survey) and from the Federal Reporting Data System, which addresses supplies delivering water with fluoride levels that exceed the current MCL. Based on the data

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from the Federal surveys and FRDS, it is estimated that over 91% of the groundwater supplies and 93% of the surface water supplies in the U.S. have fluoride present at levels of 1.0 mg/l or less. Of the 4,214 groundwater supplies estimated to have levels above 1.0 mg/l, most (2,890) fall in the range of approximately 1.0-2.0 mg/l. An estimated 276 groundwater supplies have levels above 4.0 mg/l and 16 are estimated to have levels > 8.0 mg/l. Of the 726 surface water supplies estimated to have levels > 1.0 mg/l, 704 are expected to have levels in the 1.0-2.0 mg/l range. Only 6 surface water supplies are estimated to have fluoride levels above 4.0 mg/l, and only 2 are estimated to have levels above 8.0 mg/l. Most of the groundwater and surface water supplies with levels above 2.0 mg/l are small or medium sized supplies.

The predominant sources of fluoride intake by humans are drinking water and food. For the adult male exposed to typical drinking water levels of about 1.0 mg/l, drinking water accounts for 72-91% of total intake, with the remainder contributed from food. For children 5-13 years old, drinking water at 1.0 mg/l provides 64-97% of total fluoride intake. For a newborn formula-fed infant, drinking water is the only significant source of fluoride.

INTRODUCTION

This report addresses the occurrence of fluoride in drinking water, food, and air. It has been prepared by EPA's Office of Drinking Water in connection with the assessment of existing National Interim Primary Drinking Water Regulations.

The National Interim Primary MCL for fluoride was promulgated in December 1975 (40 FR 59566) under the authority of the Safe Drinking Water Act and became effective in June 1977. The fluoride MCL applies to the approximately 60,000 community water supplies currently serving an estimated 200 million people in the United States. The current MCL for fluoride was derived from the 1962 U.S. Public Health Services Standard (27 FR 2152), which, in turn, was developed from the 1943-1946 standards.

The 1962 PHS standards addressed not only the maximum allowable fluoride levels to protect the public against excessive fluoride intake that would lead to adverse effects such as dental and skeletal fluorosis, but also the optimum level of fluoride in water known to protect against dental caries in children. The optimum level was established by the PHS to be dependent upon the annual average of the maximum daily air temperature for the location of the water supply. Air temperature was used as an indicator of daily drinking water consumption to compensate for increased water intake, especially among children, in warmer climates. The PHS standards stated that when fluoride was added to water, the average concentration should be between the lower and upper limits; when fluoride occurred naturally, the concentration should not average more than the upper limit. The PHS standards stated that fluoride concentrations averaging twice the optimum level would constitute grounds for rejection of the water supply.

The National Interim Primary Drinking Water standard addresses only the protection of the public from potential adverse effects of fluoride. The MCL was set in accordance with the 1962 PHS standards, that is, at two times the Optimum Level. Table 1 presents the current MCL and the PHS-recommended Optimum Levels.*

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*It should be noted that the accuracy of the relationship between air temperature and drinking water consumption by children has been questioned and that the Revised Regulations will probably not include the temperature dependence.

Table 1. Current Fluoride MCL and Optimum Levels for Various Temperature Ranges (in mg/liter)

Annual average of the maximum daily air temperature °F (°C)	Current MCL	Optimum level (range)
$\leq$ 53.7 ($\leq$ 12.0)	2.4	1.2 (0.9-1.7)
53.8-58.3 (12.1-14.6)	2.2	1.1 (0.8-1.5)
58.4-63.8 (21.5-26.2)	2.0	1.0 (0.8-1.3)
63.9-70.6 (21.5-26.2)	1.8	0.9 (0.7-1.2)
70.7-79.2 (21.5-26.2)	1.6	0.8 (0.7-1.0)
79.3-90.5 (26.3-32.5)	1.4	0.7 (0.6-0.8)

The evaluation of the occurrence of fluoride in drinking water, food, and air presented in this report is intended to support EPA's assessment of this substance in two principal areas. As input to the health risk assessment of fluoride, this report provides an estimate of the number of individuals in the United States exposed to various levels of fluoride in drinking water from public water supplies. Information on dietary intake and respiratory intake from ambient air is also provided for perspective and is used to evaluate the relative contributions of the three sources, particularly of drinking water, to the total dose received by individuals. While it is recognized that some individuals may be exposed to fluoride from other sources, such as occupational settings or the use of particular consumer products, this analysis is limited to drinking water, food, and air because they are the exposure routes common to all individuals.

In addition to serving as input to the health assessment, this report is also intended to support EPA efforts to estimate the economic impact of the regulatory and treatment alternatives being considered. To aid in that effort, estimates are provided in this report of the distribution of fluoride levels in public water supplies of various water source and system size categories.

An extensive body of published and unpublished information is available on the occurrence of fluoride, dating back to 1940 and in some cases earlier. This report focuses on information published since about 1965. The published information was supplemented by direct contacts with individuals at EPA, the National Institute of Dental Research, and the Center for Disease Control to obtain historical perspectives and additional information. This report is not intended to be an exhaustive, comprehensive review of all existing data on the occurrence of fluoride and human exposure to it. It does, however, present the most current and representative information available for understanding the occurrence of fluorides in drinking water, food, and air, and for assessing the importance of drinking water as a route of human exposure.

1. SOURCES OF FLUORIDE

Fluorine, F, has atomic number 9, atomic weight 19.00, and is the lightest member of the halogen family of elements (Group VII of the periodic table). It has a valence of -1 in all naturally occurring compounds. It has no isotopes. Fluorine is the most electronegative of the elements and is so highly reactive that it is almost never encountered naturally in its elemental gaseous form (F_2), but only in combined forms (including ionic and covalent, organic and inorganic). Fluoride is the generic term that is used in referring to fluorine in its combined forms.

Fluorides are ubiquitous in the human environment (NAS 1980). The occurrence of fluoride in drinking water, food, and air is influenced both by natural processes and human activities. This chapter on the sources of fluoride has three parts: The first presents a general description of the presence of fluoride in mineral deposits, soils, surface water, groundwater, the atmosphere, plants, and animals. The second part describes industrial activities that are potential as sources of fluoride to the environment. The third part provides a general description of the various routes by which fluoride moves between air, water, soil, and the biosphere as a result of natural processes and human activities. Subsequent chapters provide more quantitative data on the occurrence of fluoride in drinking water, ambient air, and food and the relative contributions of each to the daily human intake of fluoride.

1.1 NATURAL SOURCES

1.1.1 Mineral Deposits

Fluoride is relatively abundant in the earth's crust, averaging about 650 ppm by weight and ranking 13th among the elements in terrestial abundance (Shawe et al. 1976, Kirk-Othmer 1980). Fluoride minerals occur in a wide variety of geologic environments and forms. There are two major fluoride minerals: fluorspar, which contains fluorite (CaF_2), and fluorapatite, the phosphate mineral of phosphate rock [$Ca_{10}(PO_4,CO_3)_6F_{2-3}$].

The largest reserves of fluoride are found as the mineral fluorapetite in phosphate rock deposits. According to the U.S. Bureau of Mines (as cited in Kirk-Othmer 1980), the total world reserves of fluoride in phosphate rock were estimated to be 341 x 10^3 metric tons in 1977, with 28 x 10^3 metric tons in the United States. However, because the fluoride content of phosphate rock is only 3-4% by weight, its recovery is very difficult. Fluorspar, which has about 17% fluoride by weight, is the primary commercial source of fluoride. (Mined fluorspar contains about 35% fluorite, which is 48.9% fluoride.) Total 1977 world reserves of fluoride in fluorspar were 69.8 x 10^3 metric tons, with about 15 x 10^3 metric tons in the United States. Figure 1 shows the distribution of fluorspar deposits in the United States. Shawe et al. (1976) indicated that over 25% of the fluorspar mined in the United States is in the Illinois-Kentucky district; Colorado and New Mexico rank third and fourth in total fluorspar production. Other important production areas are in Idaho and Montana.

A map similar to that in Figure 1 for fluorapetite in phosphate rock deposits in the United States was not available. Economic phosphate rock deposits in the United States are primarily in the southeastern states, with some in the western states. In 1970, Florida and North Carolina were responsible for almost 80% of the phosphate rock production, Tennessee for 8%, and the western states of Montana, Wyoming, Idaho, Utah, and California for 13% combined. The amount of fluoride produced from phosphate rock is unknown, but fluoride production from this source is reportedly expected to become more prevalent in the future, since it is estimated that worldwide fluoride demand could deplete the known fluorspar reserves by the end of this century.

Other fluoride minerals include topaz [$AlSiO_4(OH,F)_2$], cryolite (Na_3AlF_6), sellaite (MgF_2), villaumite (NaF), bastnaesite [$(Ce,La)(CO_3)(F)$], and fluorine hydrosilicates [$n\ Mg(OH,F)_2 \cdot 4\ MgSiO_4 + m\ TiO_2 \cdot 4\ Mg_2TiO_2$]. Of these, topaz (in South Carolina, Colorado, and possibly New Mexico) and bastnaesite (in California) could become commercial fluoride sources.

1.1.2 Rocks and Soils

Fluoride is a common constituent of most rocks and soils. Table 2 shows the fluoride content of various igneous and sedimentary rocks. Fleischer et al. (1974) and Shawe et al. (1976) indicate that fluoride tends to occur in higher concentrations in alkalic rocks and in highly siliceous rocks, although some inverse silica-fluoride relationships have been observed. Igneous rocks with high fluoride contents tend to occur in the same two major geographic

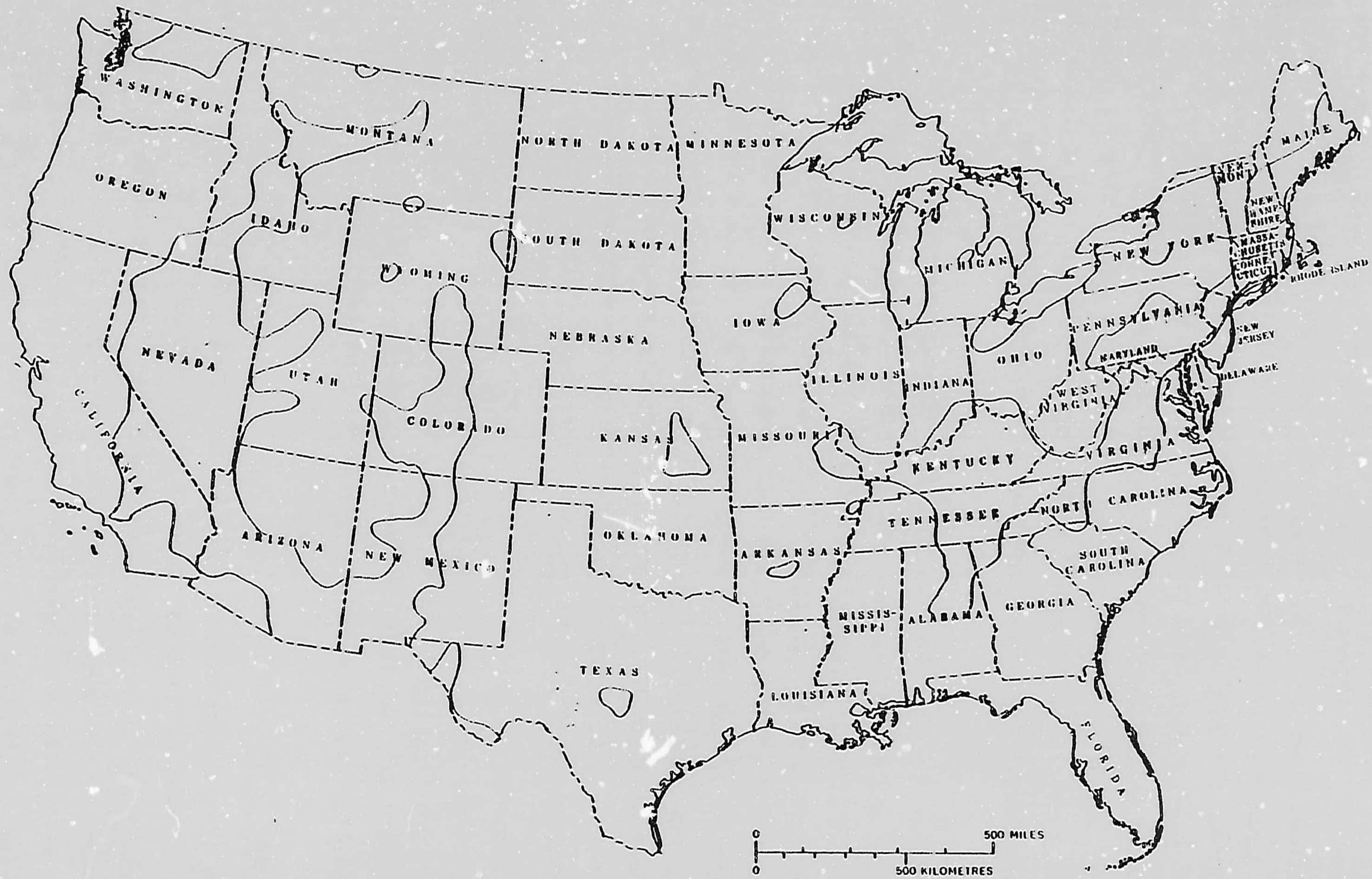

Figure 1. Distribution of fluorite deposits in the United States.

Source: Shawe et al. 1976

Table 2. Fluoride Content of Igneous and Sedimentary Rocks

Rock Type	Fluoride Content (ppm) Range	Average
Igneous		
Basalts and gabbros	20-1,100	400
Silicic rocks, granites	0-2,700	750
Alkalic rocks	200-2,200	950
Sedimentary		
Limestones and dolomites	0-1,210	230
Sandstones	10-880	180
Shales	10-7,600	800
Oceanic sediments	100-1,600	730
Volcanic ash	10-2,900	750
Soils	10-7,000	280

Source: Fleischer et al. 1974

areas shown in Figure 1 for fluorspar deposits, that is, the area extending from southern Maine southwestward to Alabama and the U-shaped area in the western states of Colorado, New Mexico, Arizona, southern California, Nevada, western Utah, Idaho, and southwestern Montana. The major sedimentary rocks having fluoride are found in the "phosphoria formation" of Idaho-Montana-Wyoming-northern Utah and in the Illinois-Kentucky district.

The natural presence of fluoride in soil is thought to be due primarily to the soil's geologic parent materials and, to a much lesser extent, to the fallout of ancient volcanic activity (Shacklette et al. 1974). Samples of soils taken at a depth of 8 inches from 911 sites located about 50 miles apart from one another throughout the United States (exclusive of Hawaii and Alaska) showed a geometric mean concentration of 180 ppm. The map and histogram in Figure 2 from Shacklette et al. (1974) show details of the results of that survey. The western United States (i.e., west of the 97th meridian) showed a geometric mean of 250 ppm, while for the eastern United States this value was 115 ppm. The higher levels in the western states tended to be associated with the central and north central Cordilleran Mountain region and probably reflect the higher fluoride concentrations in the rock types from which the soils were derived. Similar higher than average levels in the eastern states were observed in the Appalachian Highland and Ozark regions. In contrast, soils from the Atlantic Coastal Plain (southeastern area) that largely originated

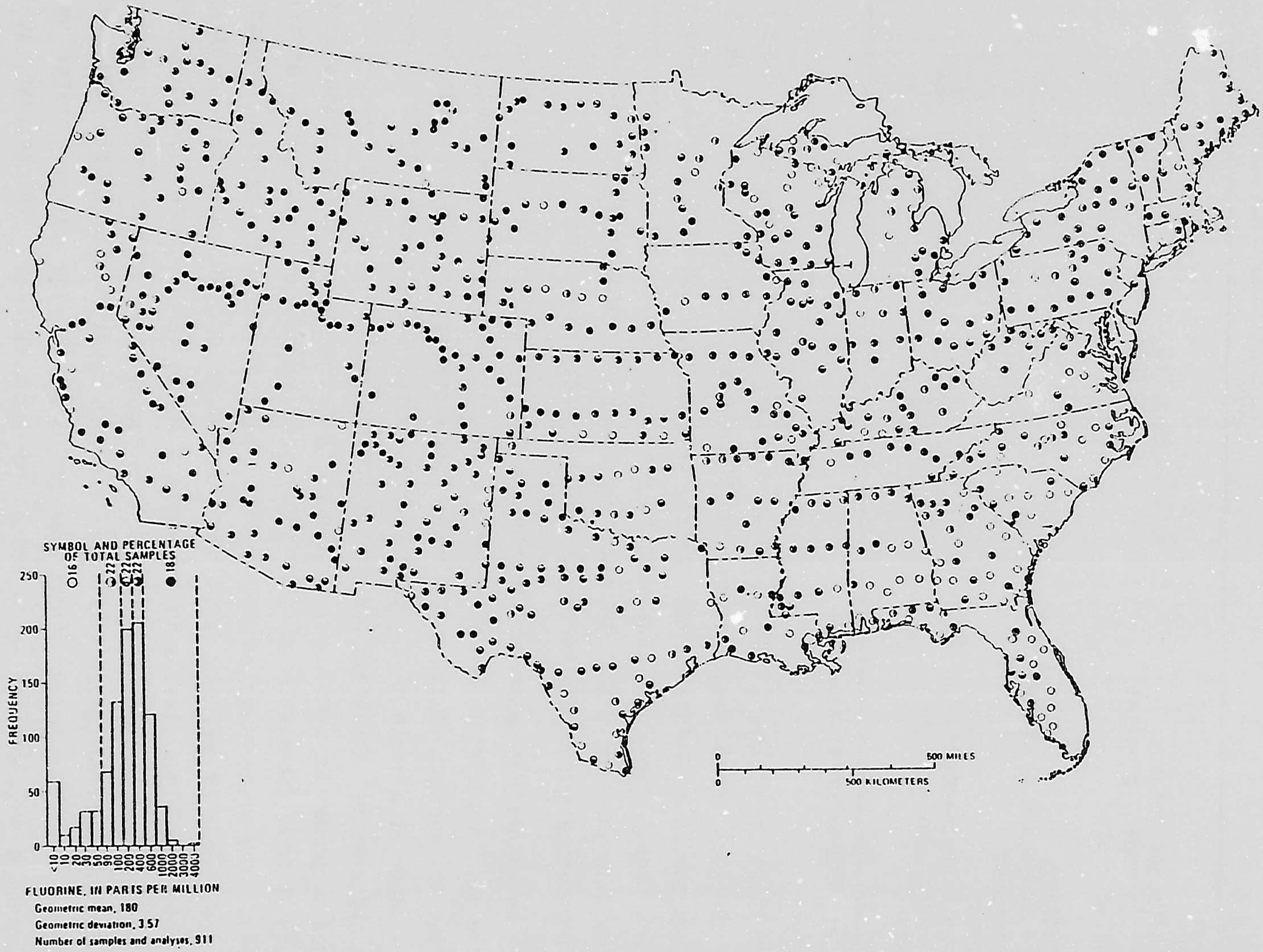

Figure 2. Fluoride concentrations in soil in the United States.
Source: Shacklette et al. 1974

from highly weathered marine deposits contain low fluoride levels. Generally, high levels of fluoride in soil samples were associated with local fluoride-rich mineral deposits and, in a few cases, with industrial activity.

1.1.3 Water

Table 3, taken from Fleischer et al. (1974), shows the range and average concentration of fluoride in seawater, surface waters, and groundwaters.

Table 3. Fluoride in Waters

Water Type	Fluoride Content (mg/l) Range	Average
Seawater	--	1.2
Groundwaters from		
Granitic rocks	0.0-9	1.2
Alkalic rocks	0.7-35.1	8.7
Basaltic rocks	0.0-0.5	0.1
Limestones and dolomites	0.0-1.7	0.3
Shales and clays	0.0-2.8	0.4
Surface waters		
Rivers	0.0-6.5	0.2
Lakes	up to 1,627	--

Source: Fleischer et al. 1974

As indicated in Table 3, seawater contains about 1.2 mg/l fluoride. According to Fleischer et al. (1974), most of the fluoride that reaches the ocean precipitates in ocean sediments. It was estimated that the amount of fluoride in seawater represents only about 0.35% of the total amount eroded from land, the remaining 99.65% having precipitated, settling to marine sediments.

Surface waters generally contain less than 1 mg/l fluoride (WHO 1970), although, as indicated in Table 3, they can contain considerably higher levels (Fleischer et al. did not comment on these high levels). The average fluoride concentration in river and lake waters of North America was reported to be 0.25 mg/l (based on 101 samples) (Shawe et al. 1976). Only the Rio Grande River system was reported to be abnormally high among North American rivers, averaging 0.6 mg/l fluoride (Shawe et al. 1976).

Britton et al. (1983) reported that the average concentration of dissolved fluoride in U.S. rivers, measured at 345 stations of the National Stream Quality Accounting Network (NASQAN) in 1976, was 0.3 mg/l. Over 75% of the streams had mean levels less than 0.5 mg/l. Only 11 streams had concentrations above 1.0 mg/l; the highest mean level was 2.23 mg/l (Table 4). Except for the Peace River at Arcadia, Florida, and the Los Angeles River at Long Beach, California, the stations with mean levels exceeding 1.0 mg/l are concentrated in the southwestern states in the Rio Grande, Lower Coloardo, Texas-Gulf and Arkansas-White-Red Water Resource Regions.

Table 4. River Stations Reporting Mean Flouride Levels Exceeding 1.0 mg/l Based on the National Stream Quality Accounting Network for the 1976 Water Year

River	Location	Mean value	Standard deviation	Range
Los Olmos Creek	(near) Falfurrias, TX	2.23	1.8	0.3-6.3
Gila River	(above) Gillespie Dam, AZ	1.89	0.61	0.5-2.8
San Pedro River	Winkelman, AZ	1.72	0.8	0.7-2.4
Los Angeles River	Long Beach, CA	1.71	3.87	0.4-14
Gila River	Calva, AZ	1.62	0.14	1.4-1.8
Beaver River	(near) Guymon, OK	1.6	0.35	1.0-1.9
Rio Grande	(near) Langtry, TX	1.54	0.3	1.0-2.2
Canadian River	(near) Canadian, TX	1.37	0.36	0.7-1.9
Peace River	Arcadia, FL	1.3	0.38	0.8-1.7
Bill Williams River	(near) Planet, AZ	1.19	0.38	0.5-1.5
Gila River	Kelvin, AZ	1.06	0.24	0.5-1.3
Beaver River	Adamsville, UT	1.02	0.4	0.6-1.8

Source: Britton et al. (1983)

As indicated in Table 3, the average fluoride content of groundwater ranges from 0.1 mg/l to more than 8 mg/l, depending upon the type of rock with which it is associated. A more expansive list of fluoride concentrations in groundwater associated with various types of rocks and water sources is shown in Table 5. Figure 3 depicts the fluoride content (maximum reported values) of U.S. groundwaters.

Table 5. Fluoride Content in Groundwaters Associated with Various Rocks and Water Sources

Rock or Water Type	Fluoride, Average in ppm (number of samples in parentheses)
Granite, rhyolite, etc.	0.9 (14)
Gabbro, basalt, ultramafic	0.2 (12)
Andesite, diorite, syenite	0.1 (4)
Sandstone, arkose, graywacke	0.4 (16)
Siltstone, clay, shale	0.6 (18)
Limestone	0.3 (14)
Dolomite	0.6 (5)
Other sedimentary rocks	0.4 (4)
Quartzite, marble	0.2 (7)
Other metamorphic rocks	0.3[a] (13)
Unconsolidated sand and gravel	0.6[b] (16)
Sodium chloride connate waters	3.1 (5)
Sodium and calcium chloride connate waters	2.0 (3)
Sulfate-bicarbonate connate waters	4.5 (7)
Spring waters similar to sodium chloride connate waters	2.1 (5)
Spring waters similar to sodium and calcium chloride connate waters	1.3 (6)
Thermal waters, volcano-associated geysers	6.1[c] (13)
Thermal sodium chloride-bicarbonate volcano-associated waters (nongeyser)	4.1[d] (6)
Volcano-associated acid sulfate-chloride springs	28.4[e] (3)
Volcano-associated acid sulfate springs	0.6 (6)
Sodium bicarbonate-boron spring waters	0.5 (5)
Thermal waters associated with epithermal mineral deposits	5.4 (11)
Nonthermal saline acid waters from mines, etc.	2.3 (3)
Travertine-depositing spring waters	4.2 (5)
Thermal waters probably meteoric in origin	0.3 (3)
Saline waters associated with salt deposits, etc.	7.7 (8)

[a]Excludes one water sample containing 4.0 ppm fluoride collected from hornblende gneiss, Transvall, Republic of South Africa.

[b]Excludes one water sample containing 24 ppm fluoride collected from lake beds, Bruneau, Idaho.

[c]Five waters associated with rhyolite, dacite, and other silicic volcanic rocks average 10.0 ppm fluoride; seven waters associated with andesite and basalt average 3.4 ppm fluoride.

[d]Four waters associated with rhyolite average 5.8 ppm fluoride; two waters associated with andesite and basalt average 0.8 ppm fluoride.

[e]Excludes one strongly acid hot-spring water sample containing 806 ppm fluoride collected from White Island (andesite volcano), New Zealand.

Source: Shawe et al. 1976

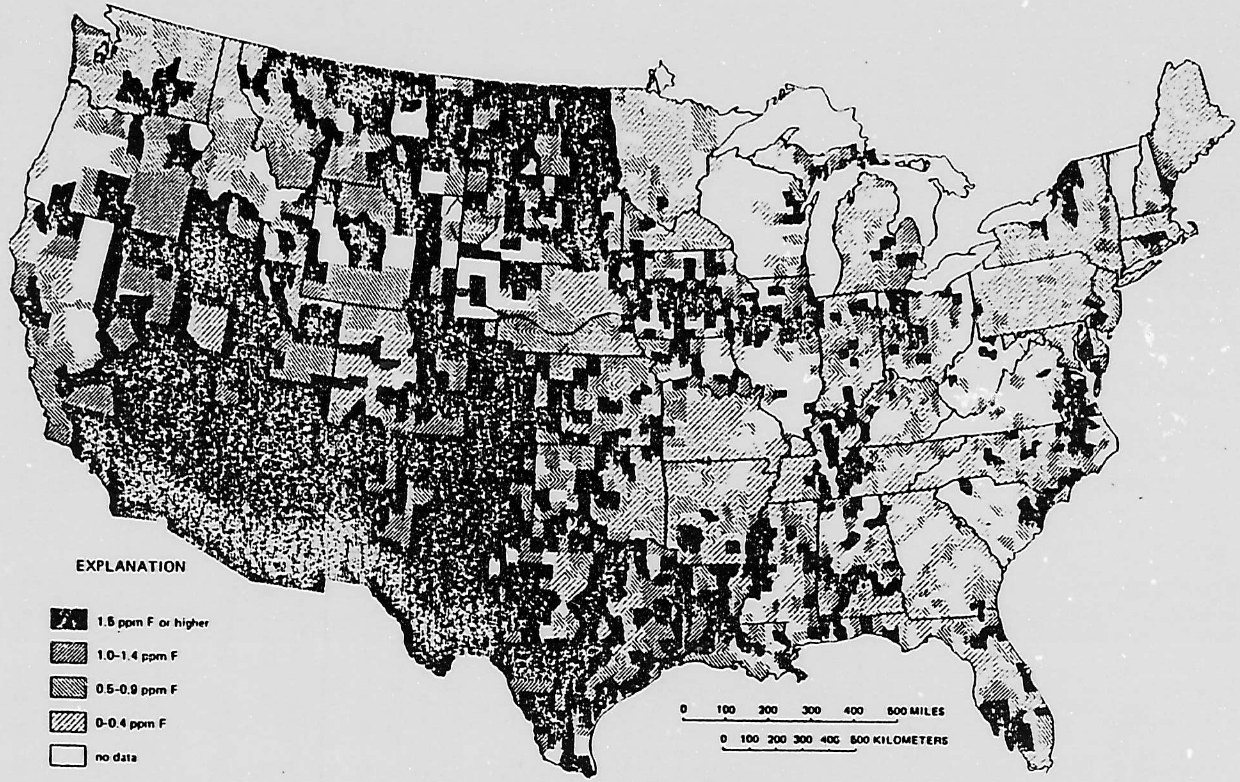

Figure 3. Fluoride content in groundwater, 1972 (maximum reported value for each county).

Source: Fleischer et al. 1974

Using the data presented in Table 5, Shawe et al. (1976) indicated that the average fluoride content of groundwater from all types of rock is about 0.4 mg/l. Higher concentrations were reported for connate waters (2.7 mg/l), and for thermal waters associated with volcanoes and epithermal mineral deposits (5.4 mg/l). Relatively high concentrations of fluoride are found in groundwaters associated with alkalic rocks, which, as mentioned earlier, tend to have higher fluoride levels than other rock types.

Groundwaters apparently dissolve fluoride from the rocks with which they are in contact in proportion to the abundance of fluoride in those rocks (Shawe et al. 1976). Some fluoride from the atmosphere (derived from the oceans and in part from volcanic and industrial activity) is reportedly also transferred to groundwater near the Earth's surface, although these sources are secondary to contact with fluoride-bearing rocks (Shawe et al. 1976).

The high fluoride levels in the groundwater of Virginia and North Carolina occur only in areas where granites are known to contain fluorite (Shawe et al. 1976). High groundwater concentrations in southern Arizona tend to coincide with areas having volcanic rocks with high fluorite content. In general, the relatively high levels of fluoride in groundwaters of the southwestern and western states tend to be dispersed around the geographic distribution of major fluorite mineral deposits (see Figure 1), although this association does not seem to hold for the eastern United States (Shawe et al. 1976). The high levels of fluoride in the groundwaters in the area that extends from northwestern Ohio westward through Iowa and then northwestward through the Dakotas (which is not considered as an area of major fluorite mineral deposits) were suggested to correlate with the glacial materials that are known to underlie this geographic region (Shawe et al. 1976).

1.1.4 Atmosphere

Gaseous and particulate fluoride compounds enter the atmosphere naturally from active volcanoes and fumaroles (vents in volcanic areas) (NAS 1980). Fleischer et al. (1974) noted that volcanic gases are rich in fluorides, mainly as hydrogen fluoride (HF), but also as silicon tetrafluoride (SiF_4) and fluorosilic acid (H_2SiF_6). Some fluorine gas (F_2) is also reportedly present, as well as a number of other fluoride compounds. Table 6 shows data on the hydrogen fluoride content of "active" gases from several volcanoes. Table 7 shows the hydrogen fluoride content of fumarole condensates from several volcanoes.

Table 6. Hydrogen Fluoride Content of "Active" Gases from Several Volcanoes

Volcano	Rock Type	HF in Weight % (number of samples in parentheses)
Hekla, Iceland	Basalt	Trace (2)
Kliuchevskii, Kamchatka	Basalt	1.6 (7)
Aso caldera, Kyushu	Basaltic andesite	0.4 (1)
Vesuvius, Italy	Tephritic leucitite	0.4 (1)
Showa-Shinzan, Hokkaido	Hypersthene dacite	2.5 (11)
Katmai, Alaska	Rhyolite	21.0 (9)

Source: Shawe et al. 1970

Table 7. Hydrogen Fluoride Content of Fumarole Condensates from Several Volcanoes

Volcano	Rock Type	HF, in mg/liter of Water (number of samples in parentheses)
Hekla, Iceland	Basalt	13.0 (1)
Santa Maria, Guatemala	Andesite	20.4 (1)
Sheveluch, Kamchatka	Andesite	38.0 (4)
White Island, New Zealand	Hypersthene andesite	69.0 (7)

Source: Shawe et al. 1976

The entrainment of soil particles and vaporization and aerosol formation from oceans, lakes, and rivers also serve as natural sources of atmospheric fluorides (NAS 1980).

No information was found on quantitative estimates of the source strengths of natural sources of fluorides, nor on the levels of fluorides in the atmosphere resulting from these natural processes. However, from the information discussed in Chapter 4, it appears that natural background levels of fluoride over the continental United States are less than 0.05 ug/m^3 (0.0625 ppb).

1.1.5 Plants and Animals

Although low concentrations of fluoride have been reported to stimulate plant growth, fluoride is not considered to be an essential element for plants. High fluoride levels in air and soil are toxic to plants; generally plants seem to be more sensitive to high air levels than they are to high soil or water levels. Shacklette et al. (1974) noted that injury to plants from fluoride in soil is controlled more by soil type, pH, calcium, and phosphorus levels -- all of which affect fluoride solubility -- than by fluoride levels themselves.

Most plants contain fluoride at concentrations of 5-10 ppm (dry weight), although the range is quite broad (< 1-300 ppm) (Fleischer et al. 1974). Most plants absorb very little fluoride from the soil, even when the soil fluoride level is naturally high or is artificially increased by the addition of fluoride chemicals (NAS 1980). Some plants are known to accumulate fluoride. Notable among these are tea (400-760 ppm), camellia (620 ppm), and elderberry (3,200 ppm) (all dry weight). Commercial tea leaves contain about 100 ppm fluoride, 90% of which is extracted by hot water. Among lower plants, brown algae are reported to have 4.5 ppm fluoride and angiosperms 0.5 ppm (Bowen 1966).

The reported concentrations (dry weight) of fluoride in animal tissues (unspecified) of various species are as follows (Bowen 1966):

Species	Concentration (ppm, dry weight)
Molluscs	2
Crustaceans	2
Fish	1,400
Mammals	150-500

Mammals, including humans (as will be discussed in more detail in Chapter 5), take up fluoride primarily from the ingestion of food and water, and, to a much lesser extent, from air. An estimated 96-99% of the fluoride in humans and animals is found in bones and teeth. Table 8 presents data on the presence of fluoride in specific mammalian tissues (Bowen 1966):

Table 8. Fluoride Concentrations in Mammalian Tissue

Mammalian Tissue	Fluoride
Bone	1,500 ppm (dry weight)
Brain	2 ppm
Heart	2 ppm
Kidney	3 ppm
Liver	4 ppm
Lung	2.4 ppm
Muscle	5.0 ppm
Blood	0.36 mg/l
Plasma	0.28 mg/l
Red cells	0.43 mg/l

Source: Bowen 1966

1.2 ANTHROPOGENIC SOURCES

As indicated in the discussion of fluoride minerals (Section 1.1.1), fluorspar is the primary commercial source of fluoride. According to Kirk-Othmer (1980), the U.S. production of fluorspar in 1972-1978 was 118,000-225,000 metric tons, mostly in the Illinois-Kentucky region. However, 80% of the U.S. consumption of fluorspar was from imported sources, mostly from Mexico.

About 50% of total U.S. consumption of fluorspar is in the steel industry, where it is used as a flux in the open-hearth process. Most of the remaining fluorspar is converted to hydrogen fluoride by treatment with sulfuric acid. About 30% of the hydrogen fluoride is consumed in the production of aluminum fluorides and synthetic cryolite for the Hall process of aluminum refining. About 40% of hydrogen fluoride consumption is for the production of chlorofluorocarbons, used as refrigerants, solvents, aerosol propellants, and in plastics. Their use as propellants has recently been significantly curtailed in the United States because of the concern about stratospheric ozone depletion. The remainder of hydrogen fluoride consumption is for stainless steel pickling, uranium enrichment, fluorine production, and the manufacture of various organic and inorganic fluorine-containing compounds, including agricultural and industrial insecticides and rodenticides, drugs and pharmaceuticals, and as a drinking water additive.

Fluorides are known to be released to the environment as a result of industrial and other human activities. Some of these involve processes in which fluorides are produced or used intentionally; in others it is a by-product of fluoride's presence in a material being used for other purposes. The primary concern historically has been with the emission of gaseous and particulate fluorides into the atmosphere from aluminum reduction plants, phosphorus and phosphate fertilizer processors, steel mills, coal burning operations, brick and tile manufacturers, and other less significant sources. Fluoride emissions to the air from these activities have in the past caused severe damage to vegetation and animals, notably to cattle feeding on contaminated forage crops. The estimated total fluoride emissions from major industrial sources in 1968 are shown in Table 9 (NAS 1980). No data more

Table 9. Estimated Total Fluoride Emissions from Major Industrial Sources in the United States in 1968[a]

Source	Atmospheric Emissions (tons/year)
Manufacture of normal superphosphate fertilizer	9,700
Manufacture of wet-process phosphoric acid	3,000
Manufacture of triple superphosphate fertilizer	300
Manufacture of diammonium phosphate fertilizer	100
Manufacture of elemental phosphorus	5,500
Manufacture of phosphate animal feed	100
Manufacture of aluminum	16,000
Manufacture of steel (open-hearth furnace)	16,800
Manufacture of steel (basic-oxygen furnace)	8,400
Manufacture of steel (electric furnace)	14,900
Welding operations	2,700
Nonferrous-metal foundries	4,000
Manufacture of brick and tile products	18,500
Manufacture of glass and frit	2,700
Combustion of coal	16,000
Total	118,700

[a]Data from U.S. Department of Health, Education, and Welfare

Source: NAS 1980

recent than these were found for atmospheric emissions. Standards of performance established under the Clean Air Act (40 CFR Part 60, Subparts S-X), however, now limit the amount of fluoride that may be emitted to the air from primary aluminum reduction plants, and the phosphate fertilizer industry (plants producing wet-process phosphoric acid, superphosphoric acid, diammonium phosphate, and triple superphosphate, and granular triple superphosphate storage facilities).

The same industries listed in Table 9 as sources of atmospheric fluorides also contribute fluoride to surface waters in their waste streams, as do other industrial operations such as electroplating (Fleischer et al. 1974). However, no information was found on the magnitude of fluoride occurrence in wastewater in general or for specific industries.

No information was found on the extent to which fluorides are found in solid wastes or in liquid wastes treated by deep-well injection.

1.3 SUMMARY OF SOURCES OF FLUORIDES IN THE ENVIRONMENT

The preceding sections of this chapter addressed the natural and human-influenced sources of fluorides in the environment. Figure 4 (NAS 1980) provides a general, if somewhat simplified, depiction of the major processes involved in the environmental transfer of fluoride.

To summarize, fluorides are natural constituents of certain rock and mineral formations in the earth's crust. Rocks and minerals appear to be the primary source of the fluoride found in soils, although deposition and precipitation of airborne particles, the application of fluoride-containing fertilizers, and plant and animals wastes also serve as sources. Fluorides enter groundwater primarily by leaching from rocks and minerals. Fluoride enters surface waters from soil runoff, industrial effluents, and deposition/precipitation of airborne particulates. Fluoride in lakes and rivers is deposited in sediments or is carried to the oceans where it is deposited in marine sediments.

Fluoride enters the air by vulcanism, entrainment of soil particles, evaporation and aerosol formation from surface waters, and industrial emissions.

In the biosphere, plants take up fluoride from soil, water, and air. Animals obtain fluoride primarily from water, and from the ingestion of plants.

No information was found on the absolute or relative magnitude of the processes that transfer and distribute fluorides throughout the environment, nor on the residence time or rate of transfer between various media. It should also be noted that the category "fluorides" is addressed very generally here, and that the transfer and residence of these chemicals among the various media also involve transformation among a great variety of specific organic and inorganic chemical forms.

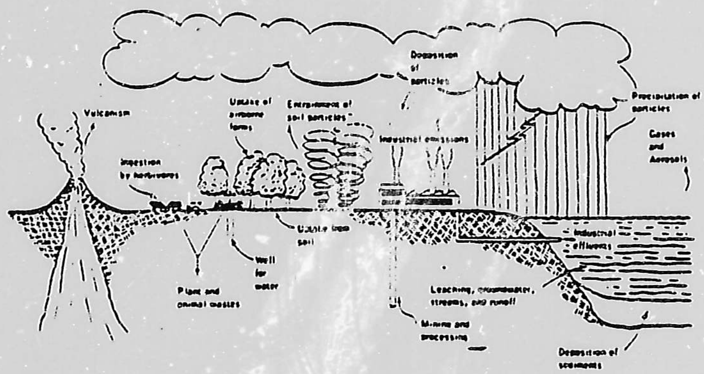

Figure 4. Environmental transfer of fluoride.

Source: NAS 1971

2. OCCURRENCE IN DRINKING WATER

The purpose of this chapter is to present information available on the levels of fluoride that have been measured in the nation's public water supplies. As discussed in the Introduction, fluoride is unique as a drinking water constitutent in that there are both a Maximum Contaminant Level (MCL) to limit fluoride levels in drinking water to protect the public from the adverse effects of fluoride, and a recommended Optimum Level for protection against dental caries. A number of drinking water systems achieve the Optimum Level by the addition of fluoride during the drinking water treatment process. As indicated in the Introduction, both the current MCL and the recommended Optimum Level are based on the ambient air temperature where the water supply system is located. As shown in Table 1, the recommended optimum level ranges from 0.7 mg/l for warmer climates to 1.2 mg/l for cooler climates; the current MCL (which is set at twice the optimum level) ranges from 1.4 mg/l to 2.4 mg/l.

As stated in the Introduction, the information presented in this report on the occurrence of fluoride in drinking water is intended to support analyses of the health risks of fluoride and of the economic impact of regulatory and treatment alternatives being considered. To achieve that, part of the initial goal of this analysis was to estimate the number of public water supplies exceeding the existing temperature-dependent MCLs.

The original approach taken to provide that estimate was to perform a separate analysis of occurrence for each area of the country falling into the various temperature ranges that determine the currently applicable fluoride MCL. However, during the course of preparing that analysis it was determined by EPA that the temperature-dependent MCL would probably be replaced in the Revised Regulations by a single nationally applicable value. Therefore, the approach taken for the analysis of fluoride occurrence presented in this report was modified to provide an estimate of the number of public water supplies nationally, independent of climate, expected to have fluoride present in several concentration ranges of interest (mg/l): $\leq$ 0.1, 0.1-1.0, > 1.0-2.0, > 2.0-3.0, > 3.0-4.0, > 4.0-5.0, > 5.0-6.0, > 6.0-7.0, and > 8.0.

2.1 REPORTED OCCURRENCE OF FLUORIDE IN PUBLIC WATER SUPPLIES

There are five major sources of information on fluoride levels in public water supplies:

- 1969 Community Water Supply Survey (1969 CWSS)
- 1978 Community Water Supply Survey (1978 CWSS)
- Rural Water Survey (RWS)
- Federal Reporting Data Systems (FRDS)
- U.S. Public Health Service Reports on Optimum Levels

The following sections describe each of those sources and present the data from each on fluoride levels in groundwater and surface water supplies.

2.1.1 1969 Community Water Supply Study (1969 CWSS)

The U.S. Public Health Service conducted a Community Water Supply Study (CWSS) in 1969 to assess the status of the nation's water supply facilities and drinking water quality (USPHS 1970a). Finished water from a total of 969 community supplies were studied, in the nine geographically distributed areas listed in Table 10. Except for the state of Vermont (in which all supplies were sampled), the study locations are standard metropolitan statistical areas (SMSAs). Water samples were reported to have been taken at various places in the distribution system of each supply studied. Of the 969 systems, 678 were groundwater supplies, 109 were surface water supplies, and the remaining 182 were mixed sources, purchased water, or of unspecified source.

Table 10. Locations Examined in the 1969 Community Water Supply Study

Region	Location	Population (in thousands)	Number of water supply systems studied
I	Vermont (entire state)	307.2	218
II	New York, New York	12,356.3	221
III	Charleston, West Virginia	229.3	30
IV	Charleston, South Carolina	251.1	22
V	Cincinnati, Ohio	1,366.0	66
VI	Kansas City, Missouri-Kansas	1,383.5	88
VII	New Orleans, Louisiana	1,085.4	26
VIII	Pueblo, Colorado	111.5	20
IX	San Bernadino-Riverside-Ontario, California	1,118.4	278
		18,203.8	969

The results of the 1969 CWSS were published in several volumes addressing each of the study areas and the national findings. The published volumes of the 1969 CWSS did not provide adequate data on the water source, population served, and fluoride levels measured for each system sampled to be useful for the national projections to be made in this study. However, a computer file with the requisite data was prepared for the 1969 CWSS from information provided by Dr. Dynager of the University of Michigan and EPA staff. A shortcoming in the 1969 CWSS data file is that no distinction can be made between those systems for which fluoride was found not to be present at the detection limit of 0.1 mg/l and those having an actual measured value of 0.1 mg/l. Therefore, all systems in the 1969 CWSS reporting a value of 0.1 mg/l were treated as though fluoride were present at that level.

Tables 11 and 12 show the reported occurrence of fluoride in groundwater and surface water supplies, respectively, sampled in the 1969 CWSS. Of the 678 groundwater supplies sampled, 628 (92.6%) had fluoride levels reported to be at or below 1.0 mg/l. Of the 50 supplies with higher levels, 34 (5.0%) were found to have fluoride between 1.0 and 2.0 mg/l, 7 (1.0%) between 2.0 and 3.0 mg/l, and 9 (1.3%) between 3.0 and 4.0 mg/l. No groundwater systems were observed in the 1969 CWSS to have fluoride levels exceeding 4.0 ug/l. Of the 16 groundwater systems exceeding 2.0 mg/l, 13 were small systems serving fewer than 500 people. The mean value of fluoride levels in groundwater systems sampled in the 1969 CWSS was 0.39 mg/l; the median was 0.17 mg/l.

In surface water, 102 (93.6%) of the 109 systems sampled had fluoride levels at or below 1.0 mg/l. Of the remaining 7 systems, 6 (5.5%) were between 1.0 and 2.0 mg/l, and 1 (0.9%) was between 2.0 and 3.0 mg/l. The mean value of fluoride levels in surface water systems sampled in the 1969 CWSS was 0.30 mg/l; the median value was 0.18 mg/l.

2.1.2 1978 Community Water Supply Survey (1978 CWSS)

The 1978 CWSS, conducted by the U.S. Environmental Protection Agency, provided data on fluoride levels in a total of 157 surface water and 345 groundwater supplies dispersed throughout the United States (USEPA 1978). The survey examined systems ranging in size from 25 people served to more than 100,000 people served. One to five samples were taken from each system and included water samples classified as raw, finished (i.e., treated water

Table 11. Reported Occurrence of Fluoride in Groundwater Systems Sampled in 1969 CWSS

System size (population served)	Number of systems studied	Number of systems with measured concentrations (mg/l) of:									
		< 0.1[a]	>0.1-1.0	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0
25-100	132	0	120	8	1	3	0	0	0	0	0
101-500	239	0	214	16	5	4	0	0	0	0	0
501-1,000	59	0	58	1	0	0	0	0	0	0	0
1,001-2,500	75	0	71	3	0	1	0	0	0	0	0
2,501-5,000	49	0	46	2	1	0	0	0	0	0	0
5,001-10,000	46	0	42	3	0	1	0	0	0	0	0
10,001-100,000	69	0	68	1	0	0	0	0	0	0	0
> 100,000	9	0	9	0	0	0	0	0	0	0	0
	678	0	628	34	7	9	0	0	0	0	0

[a]See text for discussion of supplies with values < 0.1 mg/l.

Table 12. Reported Occurrence of Fluoride in Surface Water Systems Sampled in 1969 CWSS

System size (population served)	Number of systems studied	Number of systems with measured concentrations (mg/l) of:									
		< 0.1[a]	>0.1-1.0	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0
25-100	12	0	11	0	1	0	0	0	0	0	0
101-500	19	0	19	0	0	0	0	0	0	0	0
501-1,000	14	0	14	0	0	0	0	0	0	0	0
1,001-2,500	24	0	22	2	0	0	0	0	0	0	0
2,501-5,000	10	0	9	1	0	0	0	0	0	0	0
5,001-10,000	7	0	6	1	0	0	0	0	0	0	0
10,001-100,000	15	0	14	1	0	0	0	0	0	0	0
> 100,000	8	0	7	1	0	0	0	0	0	0	0
	109	0	102	6	1	0	0	0	0	0	0

[a]See text for discussion of supplies with values < 0.1 mg/l.

sampled at the supply), and distribution (i.e., water sampled at a user's tap). For the purpose of this analysis, distribution sample data were used when available. When data for distribution samples were not available, data for finished water samples were used. Data on raw water were not included in the analysis.

Tables 13 and 14 show the reported occurrence of fluoride in groundwater and surface water systems, respectively, sampled in the 1978 CWSS. Of the 345 groundwater supplies sampled, 310 (89.9%) had fluoride levels reported to be at or below 1.0 mg/l. Of the remaining 35 systems, 23 (6.7%) were found to have fluoride between 1.0 and 2.0 mg/l, 5 (1.4%) between 2.0 and 3.0 mg/l, 3 (0.9%) between 3.0 and 4.0 mg/l, 2 (0.6%) between 4.0 and 5.0 mg/l, and 2 (0.6%) between 5.0 and 6.0 mg/l.

Of the 12 groundwater systems exceeding 2.0 mg/l, seven were small systems serving fewer than 500 people; the remaining five systems served between 500 and 2,500 people. The mean fluoride concentration in groundwater systems sampled in the 1978 CWSS was 0.58 mg/l; the median value was 0.33 mg/l.

In surface water, 134 (85.4%) of the 157 systems sampled had fluoride levels at or below 1.0 mg/l. Of the remaining 23 systems, 22 (14.0%) were between 1.0 and 2.0 mg/l, and 1 (0.6%) was reported to be at 5.0 mg/l. This latter supply was a small system serving between 101-500 people. The mean fluoride concentration in surface water systems in the 1978 CWSS was 0.69 mg/l; the median value was 0.79 mg/l.

2.1.3 Rural Water Supply

The third national survey providing data on fluoride levels in U.S. drinking water supplies is the Rural Water Survey (RWS) (USEPA 1982) conducted between 1978 and 1980 to evaluate the status of drinking water in rural America as required by Section 3 of the Safe Drinking Water Act. Although the RWS examined drinking water from over 2,000 households in rural areas for a variety of water quality parameters, samples from only 91 public water supplies were examined for fluoride levels (the sources of water for the remaining households were private wells or very small systems serving fewer than 25 people).

Table 13. Reported Occurrence of Fluoride in Groundwater Systems Studied in 1978 CWSS

System size (population served)	Number of systems studied	Number of systems with measured concentrations (mg/l) of:									
		< 0.1	≥0.1-1.0	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0
25-100	51	18	26	2	3	1	0	1	0	0	0
101-500	100	21	69	8	1	0	1	0	0	0	0
501-1,000	44	10	30	2	1	0	1	0	0	0	0
1,001-2,500	59	14	40	2	0	2	0	1	0	0	0
2,501-5,000	18	5	12	1	0	0	0	0	0	0	0
5,001-10,000	24	5	16	3	0	0	0	0	0	0	0
10,001-100,000	35	2	30	3	0	0	0	0	0	0	0
> 100,000	14	0	12	2	0	0	0	0	0	0	0
	345	75	235	23	5	3	2	2	0	0	0

Table 14. Reported Occurrence of Fluoride in Surface Water Systems Studied in 1978 CWSS

System size (population served)	Number of systems studied	Number of systems with measured concentrations (mg/l) of:									
		< 0.1	>0.1-1.0	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0
25-100	1	0	0	1	0	0	0	0	0	0	0
101-500	17	4	12	0	0	0	1	0	0	0	0
501-1,000	12	6	4	2	0	0	0	0	0	0	0
1,001-2,500	21	9	10	2	0	0	0	0	0	0	0
2,501-5,000	11	1	10	0	0	0	0	0	0	0	0
5,001-10,000	13	5	7	1	0	0	0	0	0	0	0
10,001-100,000	29	5	22	2	0	0	0	0	0	0	0
> 100,000	53	5	34	14	0	0	0	0	0	0	0
	157	35	99	22	0	0	1	0	0	0	0

A second problem with the RWS was that the number of service connections associated with water systems was reported in lieu of the actual populations served by the systems. Dr. Bruce Brower of Cornell University, who collaborated in the National Statistical Assessment of Rural Water Conditions (based on the RWS data) provided a factor to convert the data from service connections to the number of people served, based on the average number of persons per household observed in the RWS. It must be noted, however, that these population values are only approximations.

Table 15 and 16 show the reported occurrence of fluoride in groundwater and surface water systems, respectively, sampled in the Rural Water Survey. Of the 70 groundwater supplies sampled, 62 (88.6%) had fluoride levels at or below 0.1 mg/l. Of the remaining 8 systems, 6 (8.6%) had fluoride levels between 1.0 and 2.0 mg/l and 2 (2.9%) had fluoride levels between 2.0 and 3.0 mg/l. The two systems with levels between 2.0 and 3.0 mg/l served between 501 and 2,500 people. The mean value of the fluoride lvels in systems sampled in the RWS was 0.45 mg/l.

Of the 21 surface water systems sampled in the RWS, 19 (90.5%) had fluoride levels at or below 1.0 mg/l. The remaining 2 (9.5%) systems were reported to have fluoride between 1.0 and 2.0 mg/l. The mean value of fluoride in surface water systems in the RWS was 0.67 mg/l.

2.1.4 Federal Reporting Data System

The Federal Reporting Data System (FRDS) provides information on public water supplies found to be in violation of current MCLs as determined through monitoring of all supplies performed under the requirements of the National Interim Primary Drinking Water Regulations. FRDS data on supplies with fluoride violations are presented in Tables 17 and 18 for groundwater and surface water, respectively. Tables 17 and 18 also include those systems that have been granted a Variance or Exemption from the current fluoride standards as well as those systems found to be in violation of the standard through compliance monitoring. As indicated in the tables, it is currently estimated that 558 groundwater supplies and 29 surface water supplies are delivering drinking water with fluoride levels exceeding the MCL.

Because of the several different temperature-dependent standards (ranging from 1.4-2.4 mg/l) for fluoride applicable throughout the United States, the

Table 15. Reported Occurrence of Fluoride in Groundwater Systems Studied in the Rural Water Survey

System size (population served)	Number of systems studied	Number of systems with measured concentrations (mg/l) of: < 0.1	≥0.1-1.0	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0
25-100	6	1	4	1	0	0	0	0	0	0	0
101-500	11	1	9	1	0	0	0	0	0	0	0
501-1,000	15	1	12	1	1	0	0	0	0	0	0
1,001-2,500	17	2	13	1	1	0	0	0	0	0	0
2,501-5,000	10	1	7	2	0	0	0	0	0	0	0
5,001-10,000	4	1	3	0	0	0	0	0	0	0	0
10,001-100,000	5	0	5	0	0	0	0	0	0	0	0
> 100,000	2	0	2	0	0	0	0	0	0	0	0
	70	7	55	6	2	0	0	0	0	0	0

Table 16. Reported Occurrence of Fluoride in Surface Water Systems Studied in the Rural Water Survey

System size (population served)	Number of systems studied	Number of systems with measured concentrations (mg/l) of: < 0.1	>0.1-1.0	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0
25-100	0	0	0	0	0	0	0	0	0	0	0
101-500	0	0	0	0	0	0	0	0	0	0	0
501-1,000	0	0	0	0	0	0	0	0	0	0	0
1,001-2,500	2	1	1	0	0	0	0	0	0	0	0
2,501-5,000	6	3	2	1	0	0	0	0	0	0	0
5,001-10,000	2	1	1	0	0	0	0	0	0	0	0
10,001-100,000	4	1	3	0	0	0	0	0	0	0	0
> 100,000	7	1	5	1	0	0	0	0	0	0	0
	21	7	12	2	0	0	0	0	0	0	0

Table 17. National Summary of Fluoride Violations, Variances, and Exemptions in Groundwater Systems

Population served	Analytical results (range, in mg/l) Unknown	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0	Total
25-100	14	--	--	63	49	31	8	2	8	175
101-500	29	--	--	79	58	45	8	5	6	230
501-1,000	4	--	--	30	10	10	--	1	2	57
1,001-2,500	3	--	--	23	12	5	--	--	--	43
2,501-3,300	--	--	--	6	4	2	--	--	--	12
3,301-5,000	--	--	2	2	3	1	--	--	--	8
5,001-10,000	--	8	8	3	3	--	1	--	--	23
10,001-50,000	--	--	4	2	1	1	--	--	--	8
50,001-100,000	--	--	--	--	--	--	--	--	--	--
> 100,000	--	--	--	--	--	--	--	--	--	--
Unknown	--	--	--	2	--	--	--	--	--	2
National total	50	8	14	210	140	95	17	8	16	558

Source: Wentworth (1983)

Table 18. National Summary of Fluoride Violations, Variances and Exemptions in Surface Water Systems

Population served	Analytical results (range, in mg/l)									
	Unknown	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0	Total
25-100	1	--	1	--	--	1	--	--	2	5
101-500	1	1	3	1	--	--	--	--	--	6
501-1,000	--	--	1	2	--	--	--	1	--	4
1,001-2,500	1	--	3	--	--	--	--	--	--	4
2,501-3,300	--	2	--	--	--	--	--	--	--	2
3,301-5,000	--	--	1	--	-	--	1	--	--	2
5,001-10,000	--	--	1	1	1	--	--	--	--	3
10,001-50,000	--	--	1	--	--	--	--	--	--	1
50,001-100,000	--	--	1	--	--	--	--	--	--	1
> 100,000	--	--	--	--	--	--	--	--	--	--
Unknown	--	--	1	--	--	--	--	--	--	1
National total	3	3	13	4	1	1	1	1	2	29

Source: Wentworth (1983)

data in Tables 17 and 18 should be interpreted with caution. It should be noted that, for the concentration ranges of 1.0-2.0 mg/l and 2.0-3.0 mg/l, the number of systems shown is not the total number of systems in the U.S. with fluoride levels in those ranges. Rather, it is an estimate of the number of systems that by virtue of having fluoride levels in those ranges are in violation of the locally applicable fluoride standard. Other systems having fluoride concentrations in those ranges but not in violation of the local standard are not included in the FRDS data. For the concentration ranges greater than 3.0 mg/l, however, the FRDS data are considered to be a reasonably accurate reflection of the total number of systems nationally, since fluoride present at such levels would always be in violation of the MCL regardless of the local air temperature.

2.1.5 U.S. Public Health Service Reports on Optimum Levels

Two reports produced by the U.S. Public Health Service discuss fluoride in drinking water supplies, both of which deal with systems at optimal levels. The study titled National Fluoride Content of Community Water Supplies - 1969 (USPHS 1970) was conducted to identify the number and location of community water supplies in the United States that have natural fluoride levels at or above the minimum optimal level of 0.7 mg/l. This report provided the names of the communities, their populations, and the reported concentration in the community water supplies. However, no information was given on the source of the water (i.e., ground or surface) in those communities listed, nor was any information provided for communities having natural levels less than 0.7 ppm.

The results of the survey summarized in Table 19 indicate that 2,630 communities with a combined population of 8,106,435 (based on 1960 U.S. Census data) have natural fluoride levels above 0.7 mg/l. Of these, 1,517 communities (57.7%) with a combined population of 5,782,628 have drinking water supplies with natural levels in the optimum range of 0.7-1.2 mg/l. There were 1,017 communities (38.7%) with a combined population of 2,172,706 reported to have natural fluoride levels at or above 1.4 mg/l, the lowest temperature-based MCL value. Of these, 596 communities with 1,070,222 people were reported to have levels above 2.0 mg/l. Only 138 communitites were reported to have fluoride levels of 4 mg/l or more (i.e., more than twice the usual MCL) and seven communities exceeded 8 mg/l.

Table 19. Summary of 1969 PHS Study on Natural Fluoride Content of Community Water Supplies

Reported fluoride concentration (mg/l)	Number of communities	Percentage of total communities	Population	Percentage of total population	Average concentration (mg/l)	Average population of community
All levels (0.7-13.6)	2,630	100	8,106,435	100	1.55	3,082
0.7-1.2	1,517	57.7	5,782,628	71.3	0.91	3,811
$\geq$ 1.4	1,017	38.7	2,172,706	26.8	2.54	2,136
$\geq$ 2.0	596	22.7	1,070,222	13.2	3.21	1,797
$\geq$ 4.0	138	5.2	245,021	3.0	5.2	1,775

It is interesting to note that, as indicated in Table 19, the average concentration for all 2,630 communities -- which are presented in this report as those having natural fluoride levels at or above the 0.7 mg/l optimum for dental caries protection -- is 1.55 mg/l, which is in the range of MCL's. Also interesting is that 38.7% of these 2,630 communities and 26.8% of the population served are at levels above the minimum MCL of 1.4 mg/l.

The average community size in this survey was 3,082, ranging from less than 25 to one community of 313,900. Only eight communities had reported populations greater than 100,000; over 75% had fewer than 2,500. As indicated in Table 19, there is some trend toward smaller average community size for those communities with levels that exceed the current MCL. Communities reporting very high levels (e.g., 8 mg/l or more) all had populations of about 500 persons or less.

The USPHS (1970) indicated that most (62%) of the communities having natural fluoride levels of 0.7 mg/l or more were located in the states of Arizona, Colorado, Illinois, Iowa, New Mexico, Ohio, Oklahoma, South Dakota, and Texas. These same states account for about 66% of the 1,017 communities exceeding the lowest MCL of 1.4 mg/l. It is interesting to note that those states fall generally into the two areas noted in Section 1.1.3 as having high natural groundwater levels. Six states -- Delaware, Hawaii, Massachusetts, Pennsylvania, Tennessee, and Vermont -- as well as the District of Columbia were reported to have no communities with natural levels above 0.7 mg/l. Although this information indicates that there is a regional aspect to high natural fluoride levels in drinking water supplies, it is important to note that in all but one state (other than those listed above as having no natural fluoride of 0.7 mg/l or more), at least one community had 1.4 mg/l or more. (The one exception to this, New Hampshire, had one community at 1.3 mg/l.)

The second report by the U.S. Public Health Service that addresses optimum fluoride levels in drinking water supplies is the 1975 Fluoridation Census (USPHS 1977). This report is similar to the USPHS report discussed above, although it includes locations and population data for places having both natural and adjusted fluoride levels of 0.7 mg/l or more. However, this report presents no data on the concentration of fluoride in these systems. [Dr. W. Bock (1982) indicates that, like the 1969 report, the 1975 census includes all systems with levels above 0.7 mg/l, not just those in the optimum range.] Again, like the 1969 study, no information was given on water source.

Table 20. Number of Systems and Population Served by Primary Water Supply Source (by Population Category)

System size (population served)	Surface water: No. of systems	Surface water: Population (thousands)	Groundwater: No. of systems	Groundwater: Population (thousands)
25-100	1,412	55	19,632	1,033
101-500	2,383	331	15,634	3,681
501-1,000	1,341	541	4,909	3,466
1,001-2,500	1,911	1,930	4,331	6,666
2,501-3,300	514	985	881	2,440
3,301-5,000	720	1,919	1,065	4,254
5,001-10,000	912	4,626	1,159	8,078
10,001-50,000	1,306	20,771	1,101	21,530
50,001-100,000	241	10,657	84	5,289
> 100,000	218	84,541	58	12,802
Totals[a]	10,958	126,356	48,854	69,239

[a]Populations may not add to total due to rounding.

Source: Kuzmack (1982)

The 1975 Fluoridation Census reported that 9,425 places* with a total population of 105,338,343 (using 1970 U.S. Census data) have drinking water with natural or adjusted fluoride levels of 0.7 mg/l or more. Of these, 2,630 places having a population of 10,711,049 have natural fluoride levels at 0.7 mg/l or more. (The appearance of "2,630" in both the USPHS surveys is coincidental. The terms "community" used in the 1969 study and "place" used in the 1975 Census do not appear to be synonymous. Some states show significantly more "places" in the 1975 report than "communities" in the 1969 survey, while others show fewer; states showing the same number in both studies often list different locations.)

Although of limited value for this discussion of fluoride occurrence in drinking water, the 1975 Fluoridation Census does indicate a higher population receiving natural fluoride levels of 0.7 mg/l or more as compared to the 1969 study. Also, the Fluoridation Census supports the 1969 study conclusion that the nine states listed earlier are responsible for most (here 60%) of the places having natural levels of 0.7 mg/l or more.

2.2 ESTIMATED NATIONAL OCCURRENCE OF FLUORIDE IN PUBLIC DRINKING WATER SUPPLIES

There are approximately 60,000 public water supplies in the United States. As shown in Table 20, drinking water supplies fall into two major categories with respect to water source (surface water and groundwater) and into several size categories based on the number of individuals served. Section 2.1 presented the available data from Federal surveys, compliance monitoring, and two Public Health Service studies on the occurrence of fluoride in drinking water supplies. This section of the report presents estimates of the number of drinking water supplies nationally within each of the source/size categories expected to have fluoride present within various concentration ranges of interest to EPA.

It was determined that developing the national estimates required the use of both the Federal survey data and the compliance monitoring data. The

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*"Place" refers to a geographical entity listed in the Worldwide Geographical Location Codes prepared by the General Services Administration.

Public Health Service data was not useful in developing the national estimates for several reasons. Those data were for "places" or communities rather than supplies, and there was no indication as to the source of the water for those locations. The 1975 Fluoridation Census provided no quantiative data on fluoride levels in the drinking water, indicating only whether the water had natural or adjusted levels of 0.7 mg/l or more; the 1969 study addressed only communities with natural levels at or above 0.7 mg/l.

The Federal survey data together with the compliance monitoring data provide the necessary information on water source and system size for extrapolating to all public water supplies. The compliance monitoring data is believed to provide a reasonably accurate picture of those supplies in the U.S. with high fluoride levels. However, because it addresses only supplies in violation of the current MCL, the compliance monitoring data provides no information on supplies with lower levels of fluoride. The Federal survey data, which provides some information on supplies at all concentrations, is of limited value for estimating the number of systems nationally having high fluoride levels because of the small number of supplies sampled. That is, because their are relatively few supplies in the U.S. having high levels, the chances of observing them in the surveys is small. On the other hand, when a supply with a high fluoride level was observed in the surveys, extrapolating to the national level results in a questionably high national estimate. It was, therefore, decided that the compliance monitoring data would be used to describe national fluoride occurrence at the higher concentrations and the survey data would be used to estimate national fluoride occurrence at lower concentrations.

The estimates of supplies having > 3.0 mg/l are taken directly from the compliance monitoring data, since systems having levels > 3.0 would always be reported through FRDS as MCL violations. For lower levels (i.e., $\leq$ 3.0 mg/l), national estimates were calculated as follows. First, the results of the three surveys were combined as shown in Tables 21 and 22 for groundater and surface water, respectively, to determine the total number of systems sampled with values $\leq$ 3.0 ug/l, as well as the number falling within each concentration range of interest. National estimates of supplies within each concentration range were then calculated in proportion to that observed in the combined Federal survey data.

Table 21. Reported Occurrence of Fluoride in Groundwater Systems Combined Federal Survey Data

System size (population served)	Number of systems sampled[a]	Number of systems with measured concentrations (mg/l) of:			
		< 0.1[b]	$\geq$0.1-1.0	>1.0-2.0	>2.0-3.0
25-100	184	19	150	11	4
101-500	345	22	292	25	6
501-1,000	117	11	100	4	2
1,001-2,500	147	16	124	6	1
2,501-5,000	77	6	65	5	1
5,001-10,000	73	6	61	6	0
10,001-100,000	109	2	103	4	0
> 100,000	25	0	23	2	0

[a]With fluoride levels found to be $\leq$ 3.0 mg/l; total number of systems sampled including those found to have levels > 3.0 mg/l was greater.

[b]Number of systems sampled in which fluoride was not observed with a minimum quantification limit of 0.1 mg/l. That is, fluoride may be present in these systems but, if so, is at a level < 0.1 mg/l.

Table 22. Reported Occurrence of Fluoride in Surface Water Systems Combined Federal Survey Data

System size (population served)	Number of systems sampled[a]	Number of systems with measured concentrations (mg/l) of:			
		< 0.1[b]	≥0.1-1.0	>1.0-2.0	>2.0-3.0
25-100	13	0	11	1	1
101-500	35	4	31	0	0
501-1,000	26	6	18	2	0
1,001-2,500	47	10	33	4	0
2,501-5,000	27	4	21	2	0
5,001-10,000	22	6	14	2	0
10,001-100,000	48	6	39	3	0
> 100,000	68	6	46	16	0

[a]With fluoride levels found to be ≤ 3.0 mg/l; total number of systems sampled including those found to have levels > 3.0 mg/l was greater.

[b]Number of systems sampled in which fluoride was not observed with a minimum quantification limit of 0.1 mg/l. That is, fluoride may be present in these systems but, if so, is at a level < 0.1 mg/l.

For the > 2.0-3.0 mg/l range for groundwater, the estimates calculated from the Federal survey data were compared to the compliance monitoring data for each source/size category and the larger value was chosen for the national estimates. This choice was based on the recognition that the compliance monitoring data may underestimate the actual number of supplies in the > 2.0-3.0 mg/l range since not all supplies having fluoride in that range are necessarily in violation of the MCL and, therefore, would not be reported through FRDS. Higher estimates, computed from the survey data, were considered to be more conservative. On the other hand, in those instances where the compliance data showed more supplies in that range than the estimates from the survey data, the compliance data were considered more representative of actual occurrence, since the lower estimates from the survey data probably resulted from the small sample size. Specifically, for groundwater in the > 2.0-3.0 mg/l ranges, the national estimates for supplies serving 5,000 or fewer people are based on the Federal survey data and on the compliance data for supplies serving more than 5,000 persons. For surface water, all values in the > 2.0-3.0 mg/l range are from the compliance monitoring data. All estimates of groundwater and surface water supplies having ≤ 2.0 mg/l of fluoride are based on the Federal survey data.

Tables 23 and 24 present the national estimates of fluoride occurrence in public water supplies for groundwater systems and surface water systems, respectively. The estimates in Table 23 indicate that 82.8% of all public water supplies in the U.S. using groundwater have fluoride present in the range of 0.1 to 1.0 mg/l. About 8.6% of groundwater supplies (4,214 total) are estimated to have fluoride levels < 1.0 mg/l, though most of these fall in the > 1.0-2.0 mg/l range. Most of the estimated 1,324 groundwater supplies with fluoride levels exceeding 2.0 mg/l are expected to be small or medium sized supplies, while all of the supplies having very high levels (> 7.0 mg/l) are expected to be small systems serving fewer than 1,000 people.

For surface water, the majority of systems (78.5%) are also expected to have fluoride present in the 0.1-1.0 mg/l range. Of the 726 surface water supplies estimated to have levels above 1.0 mg/l, almost all (704) are estimated to be in the > 1.0-2.0 mg/l range. As in the case of groundwater, most of the surface water supplies expected to have levels exceeding 2.0 mg/l are small and medium sized systems.

Table 23. Estimated National Occurrence of Fluoride in Groundwater Systems (Number of Public Water Supplies)

Population served	Number of systems in U.S.	Concentration (mg/l)									
		<0.1[a]	0.1-1.0	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0
≤ 100	19,632	2,011	15,873	1,169	423	63	49	31	8	2	8
101-500	15,634	984	13,063	1,118	268	79	58	45	8	5	6
501-1,000	4,909	457	4,150	166	83	30	10	10	0	1	2
1,001-2,500	4,331	467	3,620	175	29	23	12	5	0	0	0
2,501-3,300	881	68	734	56	11	6	4	2	0	0	0
3,301-5,000	1,065	82	894	69	14	2	3	1	0	0	0
5,001-10,000	1,159	94	956	94	8	3	3	0	1	0	0
10,001-50,000	1,101	20	1,033	40	4	2	1	1	0	0	0
50,001-100,000	84	2	79	3	0	0	0	0	0	0	0
> 100,000	58	0	53	5	0	0	0	0	0	0	0
National total	48,854	4,185	40,455	2,890	840	208	140	95	17	8	16
Percentage of total		8.6%	82.8%	5.9%	1.7%	0.4%	0.3%	0.2%	<0.1%	<0.1%	<0.1%

[a] Includes supplies expected to have no fluoride present and supplies with fluoride present at levels below 0.1 mg/l.

Table 24. Estimated National Occurrence of Fluoride in Surface Water Systems (Number of Public Water Supplies)

Population served	Number of systems in U.S.	Concentration (mg/l)									
		<0.1[a]	0.1-1.0	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0
≤ 100	1,412	0	1,291	117	1	--	--	1	--	--	2
101-500	2,383	272	2,107	0	3	1	--	--	--	--	--
501-1,000	1,341	308	926	103	1	2	--	--	--	1	--
1,001-2,500	1,911	406	1,340	162	3	--	--	--	--	--	--
2,501-3,300	514	76	400	38	--	--	--	--	--	--	--
3,301-5,000	720	107	558	53	1	--	--	--	1	--	--
5,001-10,000	912	248	578	83	1	1	1	--	--	--	--
10,001-50,000	1,306	163	1,060	82	1	--	--	--	--	--	--
50,001-100,000	241	30	195	15	1	--	--	--	--	--	--
> 100,000	218	19	148	51	0	--	--	--	--	--	--
National total	10,958	1,629	8,603	704	12	4	1	1	1	1	2
Percentage of total		14.9%	78.5%	6.4%	0.1%	<0.1%	<0.1%	<0.1%	<0.1%	<0.1%	<0.1%

[a]Includes supplies expected to have no fluoride present and supplies with fluoride present at levels below 0.1 mg/l.

3. OCCURRENCE IN FOOD

Virtually all foods contain trace amounts of fluoride (NAS 1980). Table 25 shows some reported fluoride concentrations in various foods. Table 26 reports the fluoride content in several food categories (i.e., those used by the FDA in its market basket surveys*) taken from four areas in the United States.

Very few foods contain more than 1-2 ppm fluoride, and most contain less than 0.5 ppm (dry weight) (Underwood 1973). The notable exceptions are fish, seafoods, and tea (including camellia), which contain substantially higher levels. About 90% of the fluoride in dry tea leaves is extracted by hot water (Fleischer et al. 1974).

Table 25. Fluoride Content of Various Foods

Food	Fluoride Content (ppm)	
	WHO (1970)	NAS (1980)
Meats	0.2 - 2.0	0.01 - 7.7
Offal	2.3 - 10.1	--
Fish	5.8 - 25.9	<0.10 - 24
Shellfish	0.7 - 2.0	--
Eggs	1.2	0.00 - 2.05
Milk	0.07 - 0.22	0.04 - 0.55
Cheese	1.62	0.13 - 1.62
Butter	--	0.4 - 1.50
Tea (average, dry weight)	97.0	--
Coffee	0.2 - 1.6	0.2 - 1.6
Citrus fruits	0.03 - 0.36	0.04 - 0.36
Noncitrus fruits	0.11 - 1.32	0.02 - 1.32
Cereals and cereal products[a]	0.1 - 0.7	0.10 - 20
Vegetables and tubers	0.1 - 1.0	0.10 - 3.0
Beer and wine	0.07 - 0.24	0.15 - 0.86 (beer) 0.0 - 6.34 (wine)
Sugar	--	0.10 - 0.32

[a]Except for cottonseed, which contains 12 ppm.

"--" = no data provided.

*Fluoride is not measured by FDA in its own market basket surveys.

Table 26. Fluoride Concentrations in Market Basket Foods Purchased in Four Regions of the United States

Composite Description	Fluoride Concentrations (ppm) San Francisco (West)	Buffalo (North east)	Atlanta (South)	Kansas City (North Central)
I. Dairy products	0.05	0.05	0.07	0.05
II. Meat, fish and poultry	0.22	0.22	0.92	0.32
III. Grain and cereal products	0.34	0.39	0.41	0.29
IV. Potatoes	0.14	0.08	0.13	0.14
V. Green leafy vegetables	0.13	0.13	0.15	0.10
VI. Legumes	0.15	0.24	0.39	0.31
VII. Root vegetables	0.09	0.10	0.10	0.09
VIII. Miscellaneous vegetables and vegetable products	0.15	0.14	0.06	0.17
IX. Fruits	0.06	0.13	0.07	0.06
X. Fats and oils	0.24	0.13	0.15	0.15
XI. Sugar and adjuncts	0.21	0.24	0.32	0.35
XII. Beverages, including water	1.35	0.82	1.54	0.83

Source: NAS 1980

4. OCCURRENCE IN AMBIENT AIR

As discussed in Chapter 1, there are both natural and man-made sources of gaseous and particulate forms of fluoride to the air. Atmospheric emissions of fluorides from certain industries have resulted in serious adverse effects on vegetation and certain animals. It has been estimated that almost 120,000 tons of fluoride were emitted to the atmosphere as a result of industrial acivities in the United States in 1968 (NAS 1980). No estimates for more recent years nor for the amount of natural fluoride entering the atmosphere were found.

The most extensive data on ambient air levels of fluoride found in the literature were reported by Thompson et al. (1971), who presented information on the atmospheric concentrations of water-soluble fluorides in samples of suspended particulates. Data for more than 11,000 samples collected between 1966 and 1968 by the National Air Surveillance Network (NASN) were given. Table 27 is a summary from of these data.

Table 27. Summary of Fluoride Analyses in Air, 1966-1968

	Number of Stations	Total Number of Samples	Number of Samples with Fluoride Content (ug/m^3): <0.05[a]	0.05-0.09	0.10-0.99	>1.00	Maximum (ug/m^3)
Urban							
1966	100	2,521	2,161	152	206	2	1.89
1967	122	2,967	2,612	134	212	9	1.74
1968	147	3,687	3,287	103	290	7	1.65
1966-1968	--	9,175	8,060	389	708	18	--
Non-Urban							
1966	29	711	687	24	0	0	0.09
1967	30	729	721	5	3	0	0.16
1968	29	724	724	0	0	0	<0.05
1966-1968	--	2,164	2,132	29	3	0	--

[a]Minimum detectable level is 0.05 ug/m^3.

Source: Thompson et al. 1971

The data generally show that few urban samples and no nonurban samples exceeded 1.0 ug/m^3. The highest nonurban level recorded was 0.16 ug/m^3 in the Black Hills Forest of South Dakota in 1967; this station also recorded the highest level in 1966, 0.9 ug/m^3. No nonurban samples exceeded 0.05 ug/m^3 (the limit of detection) in 1968. More than 98% of nonurban samples during the entire 3-year period had fluoride concentrations of less than 0.05 ug/m^3.

As indicated by Table 27, 8,060 of the 9,175 urban samples (87.8%) taken during this period showed less than 0.05 ug/m^3 of fluoride. Less than 0.2% of the detectable levels exceeded 1.00 ug/m^3; none exceeded 2.0 ug/m^3. (The urban sites were characterized as being primarily commercial rather than industrial.)

Thompson et al. (1971) also presented data on about 1,700 samples taken at six urban (also primarily commercial) locations in 1967 and 1968 as part of the Continuous Air Monitoring Program (CAMP). These data, shown in Table 28, show a higher percentage of samples above the 0.05 ug/m^3 level than do the NASN urban data (30% vs. 12.2%). However, none of the CAMP concentrations exceeded 1.00 ug/m^3.

Thompson et al. (1971) indicated that reports on the levels of fluoride in plant and animal tissues in the vicinity of certain fluoride sources suggest that higher air levels can occur at some locations, although no specific data were presented. A suggested standard noted by Thompson et al. (1971) for protection of vegetation and animals was 8 ug/m^3.

A study by Chamblee et al. (1980), directed primarily at developing a more sensitive method of fluoride analysis, indicated combined gaseous and particulate levels ranging from less than 3 x 10^{-3} ug/m^3 to as much as 13.20 ug/m^3 at several locations near a phosphate mining operation in 1977. Only 16 of 56 samples (29%) had concentrations less than 0.05 ug/m^3, while 13 samples (23%) exceeded 1.00 ug/m^3; the average for these 13 samples was 4.59 ug/m^3. Two samples had concentrations greater than 10 ug/m^3 (12.22 and 13.20 ug/m^3).

Information provided from the National Aerometric Data Bank (Farrow 1982) for Arizona and North Dakota was consistent with the data reported by Thompson et al. (1971). Of 47 observations at six sites in Arizona in 1979, none exceeded the limit of detection, 0.05 ug/m^3. Of 2,837 samples taken at 37 sites in North Dakota during 1977-1978, the highest reported values were 0.30, 0.35, and 0.42 ug/m^3. Only 35 of the samples (1.2%) had levels exceeding 0.05 ug/m^3.

Table 28. Total Water-Soluble Fluoride Concentrations from CAMP Stations 1967-1968

	Number of Samples	Number of samples with fluoride content (ug/m^3)				Maximum value (ug/m^3)
		<0.05[a]	0.05-0.09	0.10-0.99	1.00-2.00	
Chicago						
1967	136	79	24	33	0	0.67
1968	151	78	44	29	0	0.55
Cincinnati						
1967	124	118	4	2	0	0.21
1968	159	134	13	12	0	0.51
Denver						
1967	111	76	18	17	0	0.2
1968	156	88	36	32	0	0.39
Philadelphia						
1967	152	119	33	0	0	0.36
1968	166	105	33	28	0	0.41
St. Louis						
1967	145	110	27	8	0	0.92
1968	155	65	36	54	0	0.46
Washington, DC						
1967	146	143	2	1	0	0.11
1968[b]	122	90	18	14	0	0.29

[a]Minimum detectable level is 0.05 ug/m^3.

[b]First 10 months.

Source: Thompson et al. 1971

5. HUMAN EXPOSURE FROM DRINKING WATER, FOOD, AND AIR

5.1 DRINKING WATER INTAKE

Section 2.2 provided estimates of the number of public drinking water supplies having fluoride present within various concentrations as a function fo water source and system size. Using those estimates together with the information shown in Table 20 on the number of people served by each source/size category of supply, the number of individuals in the U.S. receiving drinking water from public water supplies with fluoride present in the various concentration ranges of interest was calculated. These estimates are shown in Table 29.

It is estimated that over 86% of the 195,595,000 people using public water supplies are exposed to fluoride at levels of 1.0 mg/l or less; most (77.6%) are receiving water having fluoride at levels of 0.1-1.0 mg/l. Approximately 835,000 people in the U.S. are expected to be exposed to drinking water levels exceeding 2.0 mg/l. Approximately 90% of the population exposed to fluoride at levels above 2.0 ug/l receive their drinking water from groundwater sources.

Table 30 presents the estimated daily intake of fluoride from drinking water for three population groups (adult males, children 5-13 years old, and newborn formula-fed infants) as a function of the fluoride levels in drinking water. The data indicate that, on a per body weight basis, the drinking water intake of fluoride by children in the 5-13 age group is approximately 1.4 times that of the adult male, while that of newborn formula-fed infants is more than 8 times the intake of adult males. It should be noted that the drinking water intake calculations used here do not include the factor for air temperature that is allowed for in the existing EPA and PHS standards. The basis for that relationship has recently been questioned (Coniglio 1984) and the revised drinking water regulations are not expected to incorporate such a factor in the MCL.

5.2 FOOD INTAKE

Several estimates have been made of the daily dietary intake of fluoride in the United States (exclusive of drinking water). These are shown in Table 31. These estimates generally place fluoride dietary intake in the range of 0.2-0.8 mg/day.

Table 29. Estimated Population (in Thousands) Exposed to Fluoride in Drinking Water at the Indicated Concentration Ranges

System type	Number of people served in U.S. (thousands)	Number of people (thousands) exposed to fluoride at concentrations (mg/l) of:									
		<0.1	0.1-1.0	>1.0-2.0	>2.0-3.0	>3.0-4.0	>4.0-5.0	>5.0-6.0	>6.0-7.0	>7.0-8.0	>8.0
Groundwater	69,239	3,067	61,552	3,872	408.9	163.1	105.5	56.1	9.3	2.0	3.2
Surface water	126,356	13,548	90,132	22,590	71.8	6.0	5.1	0.04	2.7	0.4	0.08
Total	195,595	16,615	151,684	26,462	481	169	111	56	12	2.4	3.2
(% of total)	(100%)	(8.5%)	(77.6%)	(13.5%)	(0.2%)	(0.1%)	(<0.1%)	(<0.1%)	(<0.1%)	(<0.1%)	(<0.1%)

Table 30. Estimated Intake of Fluoride from Drinking Water

Fluoride concentration in drinking water (mg/l)	Approximate percent of population exposed to fluoride in drinking water at the indicated concentration range	Drinking water intake per individual[a] (mg/kg/day)		
		Adult males[b]	Children approximately 5-13 years old[c]	Newborn formula-fed infants[d]
0-2	99.6%	2.9×10^{-2}	4.2×10^{-2}	2.4×10^{-1}
> 2-4	0.3%	8.6×10^{-2}	1.3×10^{-1}	7.3×10^{-1}
> 4-6	< 0.1%	1.5×10^{-1}	2.1×10^{-1}	1.2
> 6-8	< 0.1%	2.0×10^{-1}	3.0×10^{-1}	1.7
> 8	<< 0.1%	2.9×10^{-1}	4.2×10^{-1}	2.4

[a]Calculations based on the midpoint of the indicated concentration range except for "> 8 mg/l) range," which uses 10 mg/l for the drinking water level; assumes 100% absorption.

[b]Calculation based on an adult male weighing 70 kg consuming 2 liters of water per day.

[c]Calculation based on a 10-year-old child weighing 33 kg consuming 1.4 liters of water per day.

[d]Calculation based on an infant weighing 3.5 kg consuming 0.8 liters of formula per day.

Table 31. Reported Daily Dietary Intake of Fluoride (exclusive of water)

Source	Category of Individual	Daily Intake (mg)
WHO (1970)	Age 1 - 3	0.027 - 0.265
	4 - 6	0.036 - 0.360
	7 - 9	0.045 - 0.450
	10 - 12	0.056 - 0.560
NAS (1980)	Adult	0.2 - 0.3
Underwood (1973)	Adult	0.3 - 0.5
Hodge and Smith (1970)	Adult	0.3 - 0.8
Singer et al. (1980)[a]	Young adult male (age 16 - 19)	0.333 (San Francisco)
		0.378 (Buffalo)
		0.587 (Atlanta)
		0.368 (Kansas City)

[a]Excludes all beverages

In contrast, Osis et al. (1974) reported a higher daily dietary intake of 1.6-1.9 mg over a 6-year period in an area with a fluoridated water supply. Kramer et al. (1974) reported fluoride dietary intakes of 1.7-3.4 mg/day in 12 cities using fluoridated water and 0.8-1.0 mg/day in four cities using non-fluoridated water. Both of these studies used a method of analysis reported by Singer and Armstrong (1965). However, Singer et al. (1980) indicated that the method used in those studies would lead to an overestimate of fluoride. While these values may not be quantitatively valid, it is interesting to note that Kramer et al. (1974) provided useful data on the correlation of dietary levels observed in the various cities with the level of fluoridation of drinking water. While no direct correlation was observed for individual cities, the mean dietary level in fluoridated cities was about three times that of the nonfluoridated cities, and the mean fluoride content of the drinking water in fluoridated cities was also about three times that of non-fluoridated cities.

5.3 RESPIRATORY INTAKE

The information on levels of fluoride in air suggest that, in general, airborne fluoride contributes little, if any, significant amount to daily intake. Assuming that an adult male inhales 23 m^3/day and absorbs 100% of inhaled fluoride, airborne fluoride present at the usual limit of detection (0.05 ug/m^3) would contribute about 1.2 ug/day to an individual's intake. This is to be compared to the estimated values of 200-800 ug/day for food and drinking water.

6. RELATIVE SOURCE CONTRIBUTION

The quantity of a substance present in the environment that is absorbed by an individual each day is the result of many personal choices and several factors over which there is little control. Where one works, lives, what one eats, and what one dose for recreation all affect daily exposure and uptake of a pollutant. People living in the same neighborhood or even in the same house can experience vastly different exposure patterns.

Table 32 shows the relative source contribution of food, air, and drinking water for fluoride intake of an adult male in the United States. The predominant sources of fluoride to the adult male in the United States are food and drinking water. As indicated in Chapter 4, typical air levels of fluoride are extremely low. Most fluoride air levels are below the limits of detection, usually 0.05 ug/m^3. Fluoride at 0.05 ug/m^3 would, with 100% absorption, contribute only 1.2 ug/day to an adult male (23 m^3/day respiration volume is assumed). For an adult male weighing 70 kg, the corresponding air dose is 1.7×10^{-5} mg/kg/day. Except where the food and drinking water doses are 0, the air contribution appears to be negligible.

The food intake shown was derived from the data presented in Section 5.2, which suggested that the daily dietary intake was 0.2-0.8 mg/day. Assuming 100% abosrption for a 70-kg adult male, these values correspond to $2.9 \times 10^{-3} - 1.1 \times 10^{-2}$.

Under the typical drinking water exposure conditions of about 1.0 mg/l, drinking water accounts for an estimated 72-91% of total fluoride intake for the adult male, with the remainder contributed from food. Where drinking water levels exceed 2 mg/l, the contribution from drinking water is generally expected to exceed 90% of total intake.

A similar comparison of relative source contribution is shown in Table 33 for children approximately 5-13 years old. As in the case of the adult male, the contribution from air is negligible relative to drinking water and food. Under typical drinking water exposure conditions of about 1.0 mg/l, drinking water accounts for 64-97% of total fluoride intake. Again, where drinking water levels exceed 2.0 mg/l, drinking water generally accounts for more than 90% of total intake.

Table 33. Estimated Intake of Fluoride from the Environment by Children Approximately 5-13 Years Old[a] (mg/kg/day)

Fluoride concentration in drinking water (mg/l)	Estimated population of children 5-13 years old exposed to indicated fluoride concentration range from public water supplies[b]	Drinking water intake per individual[c] (mg/kg/day)	Total intake per individual in mg/kg/day (% from drinking water) — Food intake per individual[d] (mg/kg/day): 1.1×10^{-3}	1.3×10^{-2}	2.4×10^{-2}
0-2	26,039,600	4.2×10^{-2}	4.3×10^{-2} (97%)	5.5×10^{-2} (76%)	6.6×10^{-2} (64%)
> 2-4	86,800	1.3×10^{-1}	1.3×10^{-1} (99%)	1.4×10^{-1} (91%)	1.5×10^{-1} (84%)
> 4-6	22,300	2.1×10^{-1}	2.1×10^{-1} (99%)	2.2×10^{-1} (94%)	2.3×10^{-1} (90%)
> 6-8	1,900	3.0×10^{-1}	3.0×10^{-1} (100%)	3.1×10^{-1} (96%)	3.2×10^{-1} (93%)
> 8	400	4.2×10^{-1}	4.2×10^{-1} (100%)	4.3×10^{-1} (97%)	4.4×10^{-1} (96%)

[a]Daily intake from air (estimated to be less than 2.3×10^{-5} mg/kg/day) considered negligible relative to food and drinking water.

[b]Based on 1981 data provided in Statistical Abstract of the United States 1982-83 showing 13.37% of the total U.S. population falling in the 5-13 years old age group.

[c]Calculation based on a 10-year-old child weighing 33 kg consuming 1.4 liters of water per day using the midpoint of the indicated concentration range except for the "> 8 mg/l range" which uses 10 mg/l for the drinking water level.

[d]Based on data showing the daily dietary intake of fluoride for children ages 4-12 to range from 0.036-0.56 mg/day and assuming a body weight of 33 kg (1.1×10^{-3} mg/kg/day = 0.036 mg/day; 1.3×10^{-2} mg/kg/day = 0.30 mg/day; 2.4×10^{-2} mg/kg/day = 0.56 mg/day).

Table 32. Estimated Intake of Fluoride from the Environment by Adult Males[a] (mg/kg/day)

Fluoride concentration in drinking water (mg/l)	Estimated percent of the population exposed to indicated fluoride concentration range from public water supplies (% of total)	Drinking water intake per individual[b] (mg/kg/day)	Total intake per individual in mg/kg/day (% from drinking water) — Food intake per individual[c] (mg/kg/day): 2.9×10^{-3}	7.1×10^{-3}	1.1×10^{-2}
0-2	99.6%	2.9×10^{-2}	3.2×10^{-2} (91%)	3.6×10^{-2} (81%)	4.0×10^{-2} (72%)
> 2-4	0.3%	8.6×10^{-2}	8.9×10^{-2} (97%)	9.3×10^{-2} (92%)	9.7×10^{-2} (89%)
> 4-6	< 0.1%	1.5×10^{-1}	1.5×10^{-1} (98%)	1.6×10^{-1} (95%)	1.6×10^{-1} (93%)
> 6-8	< 0.01%	2.0×10^{-1}	2.0×10^{-1} (99%)	2.1×10^{-1} (97%)	2.1×10^{-1} (95%)
> 8	<< 0.01%	2.9×10^{-1}	2.9×10^{-1} (99%)	3.0×10^{-1} (98%)	3.0×10^{-1} (96%)

[a]Daily intake from air (estimated to be less than 1.7×10^{-5} mg/kg/day) considered negligible relative to food and drinking water.

[b]Calculation based on an adult male weighing 70 kg consuming 2 liters of water per day and using the midpoint of the indicated concentration range except for the "> 8 mg/l range," which uses 10 mg/l for the drinking water level.

[c]Based on data showing the daily adult dietary intake of fluoride ranging from 0.2-0.8 mg/day and a 70-kg adult (2.9×10^{-3} mg/kg/day = 0.2 mg/kg/day; 7.1×10^{-3} mg/kg/day = 0.5 mg/day; 1.1×10^{-2} mg/kg/day = 0.8 mg/day).

Lastly, Table 34 shows the esimated intake of fluoride for newborn bottle-fed infants. In this case, both food and air are considered to be negligible relative to drinking water used to prepare formula.

Table 34. Estimated Inatke of Fluoride from the Environment by Newborn Formula-fed Infants[a]

Fluoride concentration in drinking water (mg/l)	Estimated population of formula-fed infants exposed to indicated fluoride concentration range from public water supplies[b]	Drinking water intake per individual[c] (mg/kg/day)
0-2	1,651,500	2.4×10^{-1}
> 2-4	5,500	7.3×10^{-1}
> 4-6	1,400	1.2
> 6-8	122	1.7
> 8	30	2.4

[a]Food is not a source for newborn formula-fed infants; intake from air (estimated to be 1.1×10^{-5} mg/kg/day) considered negligible.

[b]Based on data indicating infants under 1 year of age comprise 1.6% of the U.S. population and that 53% of infants are formula-fed, having drinking water in the formula as their primary source of fluid.

[c]Calculation based on an infant weighing 3.5 kg consuming 0.85 liter of formula per day, using the midpoint of the indicated concentration range except for "> 8 mg/l" range, which uses 10 mg/l for the drinking water level.

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